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A new Optical Technique for Investigating Surface Nanoscale Electron Physics

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6. AUTHOR(S)

Prof. Richard M. Osgood, Jr.

7. PERFORMING ORGANIZATION NAMES(S) AND ADDRESS(ES)

Columbia University
Department of Electrical Engineering
S.W. Mudd Bldg., Rm. 1322
New York, NY 10027

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13. ABSTRACT *(Maximum 200 words)*

This report describes the acquisition, set-up and use of essential equipment for studying surface nanoscale electronics and the progress in the related research projects where such equipment has been or will be used. During the past year, using this DURIP award, we installed a larger UHV chamber, implemented an auxiliary ultraviolet photoemission system (UPS), and established an electron-beam writing system. We have studied several nanostructured surface systems in stepped single-crystal surfaces and observed electron lateral 1D confinement and coherent effects using high-resolution angle-resolved two-photon photoemission. We investigated height-dependent (in nanoscale) electronic structures using sp-like surface state and image state electrons and observed distinct differences in the electron kinetics. We have been able to successfully generate patterns of dots and line structures of approximately 100nm in a variety of metal and semiconductor substrates.

14. SUBJECT TERMS	15. NUMBER IF PAGES
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DURIP Final Technical Report
DAAH04-95-1-0383: "A New Optical Technique for Investigating Surface Nanoscale Electron Physics"
Principal Investigator:
Richard M. Osgood, Jr., Columbia Radiation Laboratory

Our research efforts at Columbia Radiation Laboratory have been in the fields of interaction of ultraviolet laser radiation and metal/semiconductor surfaces and novel fabrication techniques for electronic and opto-electronic applications. This DURIP award allowed us to acquire essential equipment for our ongoing investigation in surface nanoscale electronics. The three main equipment sub-systems purchased using this fund are: a) UHV (ultrahigh vacuum) chamber which allows us to implement new diagnostic tools for surface electronics characterization and improve the existing system; b) Auger diagnostic system: we acquired i) a multi-technique hemispherical energy analyzer which can be used for various surface science applications, such as ultraviolet photoemission spectroscopy (UPS), x-ray photoemission spectroscopy (XPS), Auger electron spectroscopy (AES), and ion scattering spectroscopy (ISS); ii) a UPS light source, which can generate hard-UV mono-energetic photons (spectral lines from 11 to 48 eV), to attain an auxiliary (self-sufficient) UPS system for *in situ* analysis of surface and bulk electronic states (occupied) to provide supplementary information on the electronic structure of the surface and substrate; iii) an e-beam writing system: we purchased nanometer pattern generation system (NPGS) and accessories to convert a scanning electron microscopy (SEM) system and obtain the ability to fabricate nanoscale structures.

Our current research emphasis is to study electronic phenomena and understand quantum confinement and coherent effects at nanostructured metal surfaces using our novel nonlinear photoemission technique. The effects of surface nanostructure on electron confinement at the surface have recently attracted much attention due to the desire to engineer electronic and magnetic materials on the atomic scale. Semi-artificially, we can prepare regularly stepped surfaces with well-defined step density and orientation by the use of small-angle miscuts to the low-index single-crystal planes. Such steps are typically of nanometer height (2~3Å) and periodicity (10~30Å), and serve well as a prototype of low-dimensional (quantum wire) systems. Early studies showed markedly different electronic properties on the stepped surfaces than on flat low-index surfaces. Among them are work-function reduction, larger sticking probability for

adsorbates, and certain standing wave phenomena as detected by STM (scanning tunneling microscopy). However, these are isolated local effects and the magnitude of the electronic structure of the steps and the effectiveness of the influence of the step potential on electrons at various energy states have not been extensively studied. In our experiments, we have examined two vicinal surfaces [Cu(1 $\bar{1}$ 9) and Cu(775)]: stepped Cu(001) and Cu(111). By using various Angle-Resolved Two-Photon Photoemission (AR2PPE) techniques, we were able to probe the electronic structure as a function of distance from the surface along the surface normal and observed different electronic behaviors for the surface electrons depending on their locations with respect to the sample surface.

In initial experiments, we demonstrated the first direct observation of *lateral* superlattice effects for image-state electrons on stepped Cu(001) using wide-angle UV AR2PPE. Our results show that the lateral periodicity (~11Å) of the bare stepped surface leads to back-folding of the dispersion of energy versus parallel momentum, $k_{\parallel}$, of the image state. Further, adsorption of Na atoms (~0.01 monolayer) on the stepped surface enhances the step regularity and sharpens this characteristic dispersive behavior to the point that it can be followed to the edges of the lateral Brillouin zone formed by the step lattice. The reduction of the surface Brillouin zone by a factor of 4.5, compared to that of planar Cu(001), allows a clear observation of back-folding of the free-electron-like dispersion which is normally seen on planar surfaces, yielding an oscillatory dispersive behavior where the electron energy is a multivalued function of $k_{\parallel}$ as seen in the first two Brillouin zones of such a superlattice. The effects of the additional reciprocal lattice vector from the step lattice appear to have not been seen previously in the band structure of nanostructured surfaces. The results present a classic example of the 1D quantum nature that can be described by a simple Kronig-Penney model for electrons in a regular periodic potential. The fact that we observe an effect of nanostructuring on the electronic states encourages the development of artificial structures as tailored electronic materials.

In the second set of experiments, we performed comparative studies of the electronic structures (the n=0 *sp*-like surface state, and n=1 and 2 image states, each of which has different distances from the surface plane) on Cu(111) and stepped Cu(775) using angle-resolved *resonant* as well as non-resonant *bichromatic* or monochromatic two-photon photoemission. For Cu(775), monatomic steps are formed along [1 $\bar{1}$ 0] direction and its (111) terrace consists of 7 rows of

[$1\bar{1}0$]-oriented atoms —giving rise to an intrinsic terrace width of d_0 = 14.02Å. By tuning the photon energy of nanosecond laser pulses, resonant bichromatic two-photon excitation from the *sp* surface state (n=0) to the image-potential states (n=1,2) can be achieved at various values of electron momenta parallel to the surface ($k_{\parallel}$) and hence the energy bands of both the initial and intermediate states can be mapped out simultaneously. These image states are two-dimensional in nature and the image-state electron is free-electron-like in a plane parallel to its image plane, and hence its parabolic dispersion curve is determined by the image plane orientation, *i.e.* the dispersion band minimum occurs at the normal of the image plane. A phase-analysis calculation shows that the n=1 image state is located about 3Å above the image plane and the n=2 image-state electron exists about 12Å above the surface. Because of its close distance above the surface, the n=1 image-state electron can be expected to see the image plane formed on a local (111) terrace instead of the general plane of the (775) surface. In other words, the step terrace of over 10Å is large enough to support the image state. The measured dispersion data of the n=1 state agree well with this explanation: the maximum binding energy is at $k_{\parallel} \sim 0.12$ Å^{-1}, which corresponds to the terrace normal [111], instead of the surface normal at $k_{\parallel} = 0$, revealing that the n=1 state is oriented with the step terrace not the general surface plane of Cu(775). With regard to the n=2 state, this higher Rydberg state is much further (~12Å) above the surface and thus sees the overall, averaged surface, *i.e.* the general Cu(775) surface, as the image plane rather than the detailed step-terrace structures. Thus, its dispersion is oriented so as to align with the Cu(775) surface not the step terrace in the [111] direction, *i.e.* the dispersion minimum remains at $k_{\parallel} = 0$. These dispersion orientations of the n=0, 1, and 2 states, can be summarized as follows: the n=2 image state senses the general surface plane, the n=1 image state senses the local terrace plane, and the n=0 surface state, which exists ~ 4 Å beneath the surface, does not appear to be affected by the steps.

Angle-resolved measurements were also done at larger $k_{\parallel}$ values via monochromatic UV AR2PPE at different fixed photon energies on the stepped Cu(775). One important feature is that different sets of measurement at different photon energies all showed dispersion relations with a common oscillation period and thus electron bandfolding effects (such as described above for stepped Cu(001)) occurred at the same turning position despite different electron energies at the turning point. This behavior is clear evidence of Umklapp processes for the n=1 image-state

electrons in the step lattice on Cu(775), where electrons are localized ~3Å above the surface. The dispersion curves can be transformed in the k-space by the step reciprocal-lattice vector, which is of the magnitude $0.22Å^{-1}$. Note that such effects have not been seen by other surface electron experiments on stepped surfaces. This is another aspect that the embedded n=0 surface electron does not see the stepped structures while the n=1 image state does. The differences in the height-dependent electronic structure show that the n=1 image state is most sensitively affected by the step structure while the embedded *sp* surface state is not noticeably affected by the presence of steps, but rather determined only by the projection of the bulk band structure on the (775) surface.

With the new UHV chamber installed, we have been able to set up the newly acquired 100mm-radius hemispherical electrostatic energy analyzer and the differentially pumped UHV-compatible UV light source. With this UPS system in place, we have been able to obtain preliminary one-photon photoemission results on the occupied electronic states on Cu(111), Cu(775) and Si(111). We also implemented a time-of-flight (TOF) electron detector for fast, high-resolution data acquisition. Further improvement of the existing angle-resolved 160° spherical-sector energy analyzer will soon be undertaken. We have successfully converted our SEM into an electron-beam writing system with the purchase of the NPGS software and accessories. Using this device we have been able to pattern line structures, down to 130nm consistently with good control. This allows us to also make arrays of 90x120nm dots. These have been done on PMMA resist, on GaAs, Si and Al. Interestingly, we have also started patterning features that can be used to make optic devices, such as nanoscale corner mirrors. The patterns exhibited the sharpness needed for the application. These equipment additions have significantly augmented our capacity in the research activities in our current ARO- and JSEP-funded programs as mentioned above as well as a joint ARPA/AFOSR program by the principal investigator to develop novel fabrication techniques for damage-free nanostructures on semiconductor surfaces and masking techniques for optical devices. They will play a very important role in our next project investigating quantum well states and giant Magnetoresistance (GMR) in metal multilayer structures, incorporating novel techniques utilizing ultrafast laser technology.

List of Equipment Items Acquired

	Vendor	Cost
1. UHV Chamber	Kurt J. Lesker Co.	$ 8,417
2. Auger Diagnostic System		
a) Hemispherical Analyzer Model CL100M12	Micro Photonics	$ 58,874
b) UPS Light Source Model UVL-HI	Kurt J. Lesker Co.	$ 10,018
c) UV Film Polarizer Model TFK2-266-60	CVI Corp.	$ 1,815
3. E-beam Writing System		$ 22,000
a) Nanometer Pattern Generation System	JC Nabity Lithography System	
b) SEM Conversion Accessories	Omicron Associates First Source Int'l.	$ 1,295 $ 203
TOTAL		$102,622